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Comment on acp-2021-876

Anonymous Referee #3

Referee comment on "The impacts of marine-emitted halogens on OH radicals in East Asia during summer" by Shidong Fan and Ying Li, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-876-RC3, 2021

Review of Fan and Li, the impact of marine-emitted halogens on OH radical in East Asia during summer

General comments:

This manuscript analyzes regional model simulation results regarding the impact of tropospheric halogen chemistry (Cl, Br, and I) on the production rates and concentrations of OH over East Asia and Western Pacific during summer. Three major pathways, i.e., O3 photolysis rate, HOX photolysis, and HO2 +Y were identified and the changes were quantified per emission types (SSA, inorganic iodine, and halocarbons) and their associated processes (chemistry, radiation, and deposition). Although not proposing new processes, the authors describe the overall impact in an integrated manner and then their attribution to individual processes. The Western Pacific low-latitude region is with fairly high sea surface temperature during summer and thus potentially high impact of iodine chemistry (due to high [I-] in the surface seawater) is expected. The findings are mostly reasonable and the logical flow of the manuscript appears sound.

Nonetheless, there are several points needing clarification. First, additional fundamental information of this study is required for justification. For example, a table of chemical reactions relevant to tropospheric halogen chemistry taken into account in the simulations is needed at least in the supplement. Maps of the assumed SSA, HOI and halocarbon fluxes will also help understanding. Typical air transport patterns during July 2019 should

be described.

Second, clarification is necessary for some processes. For example, I am afraid that the impact of sea spray aerosols on J(O1D) (Fig. S5, Fig. 6a) is a bit too large. The aerosol optical depth and assumed single scattering albedo in the model need evaluation. Also I do not understand why the InorgI_chem can result in "negative" values over the Philippine Sea, although some explanation is given in lines 379-382.

Third, readability of the manuscript should be improved, as very similar figures (from Figure 2 to 7) appear from one to another. Overall, the manuscript is acceptable after major revisions, responding to the points raised above and to specific comments listed below.

Specific comments:

- Line 20. Show what are the "three" major pathways in Abstract.
- Line 22. The increased ozone deposition due to enhanced I- ion levels in the surface seawater would be rather "chemical" than physical?
- Line 41. What are the "artificial" and "mechanistic" pathways?
- Line 50. longer-lived?
- Line 51 resulting in
- Line 52. It is not box models that are to be accused. Maybe "constraining O3 levels in the model" causes the difficulty that the authors described.
- Lines 57 and 260. I believe previous CTM studies implicitly take the impact from HO2 into account, unless they constrain HO2 levels to observations.
- Line 77. What are "its extreme uncertainties"?
- Line 90. List tropospheric halogen chemistry reactions from the CB6r3m mechanism in a supplementary table. Are they identical to those involved in GEOS-Chem or other recent studies (e.g., Sherwen et al., 2016, Stone et al., 2018, Wang et al., 2021), which are cited in line 202 and compared?
- Lines 99-100. The typical air mass transportation pattern for this region during July 2019 should be described.
- Line 140. I believe these two processes are coupled in Sekiya et al. (2020).
- Table 1. One idea would be to add one column in the right to indicate in which Figure the results of the cases are studied.
- Line 242. Explain what FORM+O is.
- Section 3.1. The model performance is only checked with O3 monitoring over the continent. It would be useful to compare the simulation results also with O3 observations at Yonaguni available from http://ebas.nilu.no.

- Figures 3-7. In their figure captions, some more explanation should be included, to clarify cross linkage of the figures. For example, Figure 3 caption could include that they are breakdown of Figure 2a down to processes. Figure 4 would be the breakdown of Figure 3g, a, e, and f. etc.
- Lines 305 and 312 and Fig. S5. I am afraid that the impact of sea spray aerosols on J(O1D) and then P(OH) (close to 30% for daytime maximum) is a bit too large. The aerosol optical depth and assumed single scattering albedo in the model need evaluation.
- Line 370. What is the process that "iodine" diminishes the photolysis of O3?
- Lines 379-382. Why can the InorgI_chem result in "negative" values over the Philippine Sea? I believe that the photolysis of HOI always tend to "increase" OH and thus should have positive values. Coupling with air mass transport may produce this type of feature? Clarification is needed.
- Line 392. Rely much on the current halogen chemistry
- Line 395. The authors should explain how the debromination is included.
- Line 400. Indeed, the uptake of HOI onto the sea salt particles could significantly alter the fate of HO2 and then OH (i.e. HO2 + Y term) and most of the results in this study. This was previously studied by Kanaya et al. GRL 2002 in the Asian domain.
- Line 409. There are a series of laboratory studies examining uptake of HO2 onto sea-spray aerosol particles (Taketani et al., 2008, 2009).
- Line 413. What is the "mass interaction"?
- Throughout the manuscript: Was all P(OH) studied always at the lowest model layer? How will the results change, when studying the whole atmospheric boundary layer?

References

Kanaya, Y., Yokouchi, Y., Matsumoto, J., Nakamura, K., Kato, S., Tanimoto, H., Toyota, K., and Akimoto, H.: Implications of iodine chemistry for daytime HO2 levels at Rishiri Island, Geophys. Res. Lett., 29(8), doi:10.1029/2001GL014061, 2002.

Sekiya, T., Kanaya, Y., Sudo, K., Taketani, F., Iwamoto, Y., Aita, M. N., Yamamoto, A., and Kawamoto, K.: Global Bromine- and Iodine-Mediated Tropospheric Ozone Loss Estimated Using the CHASER Chemical Transport Model, Sola, 16, 220-227, 10.2151/sola.2020-037, 2020.

Sherwen, T., Schmidt, J. A., Evans, M. J., Carpenter, L. J., Grossmann, K., Eastham, S. D., Jacob, D. J., Dix, B., Koenig, T. K., Sinreich, R., Ortega, I., Volkamer, R., Saiz-Lopez, A., Prados-Roman, C., Mahajan, A. S., and Ordonez, C.: Global impacts of tropospheric halogens (Cl, Br, I) on oxidants and composition in GEOS-Chem, Atmospheric Chemistry and Physics, 16, 12239-12271, 10.5194/acp-16-12239-2016, 2016.

Stone, D., Sherwen, T., Evans, M. J., Vaughan, S., Ingham, T., Whalley, L. K., Edwards, P.

M., Read, K. A., Lee, J. D., Moller, S. J., Carpenter, L. J., Lewis, A. C., and Heard, D. E.: Impacts of bromine and iodine chemistry on tropospheric OH and HO2: comparing observations with box and global model perspectives, Atmospheric Chemistry and Physics, 18, 3541-3561, 10.5194/acp-18-3541-2018, 2018.

Taketani, F., Kanaya, Y., and Akimoto, H.: Heterogeneous Loss of HO2 by KCl, Synthetic sea salt and natural seawater aerosol particles", Atmos. Environ. 43, 1660–1665, 2009.

Taketani, F., Kanaya, Y., and Akimoto, H.: Kinetics of heterogeneous reaction of HO2 radical at ambient concentration levels with (NH4)2SO4 and NaCl aerosol particles, J. Phys. Chem. A, 112 (11), 2370 -2377, doi:10.1021/jp0769936, 2008.

Wang, X., Jacob, D. J., Downs, W., Zhai, S., Zhu, L., Shah, V., Holmes, C. D., Sherwen, T., Alexander, B., Evans, M. J., Eastham, S. D., Neuman, J. A., Veres, P., Koenig, T. K., Volkamer, R., Huey, L. G., Bannan, T. J., Percival, C. J., Lee, B. H., and Thornton, J. A.: Global tropospheric halogen (Cl, Br, I) chemistry and its impact on oxidants, Atmos. Chem. Phys. Discuss., 2021, 1-34, 10.5194/acp-2021-441, 2021.